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NBSIR 83-2646

Characterization of Microwave Window Materials

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U.S. DEPARTMENT OF COMMERCE
National Bureau of Standards
National Measurement Laboratory
Center for Materials Science
Inorganic Materials Division
Washington, DC 20234

May-October 1982

Interim Report

Issued January 1983

Task Code: NggQAXAH/ Missile Systems/ X-Ray Hardness
Work Unit: 00043/ Materials Characterization

Prepared for

QC — Defense Nuclear Agency
100 Washington, DC 20300

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CHARACTERIZATION OF MICROWAVE WINDOW MATERIALS

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U.S. DEPARTMENT OF COMMERCE, Malcolm Baldrige, *Secretary*
NATIONAL BUREAU OF STANDARDS, Ernest Ambler, *Director*

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Introduction

Since February 1982, NBS has been engaged in a project concerned with the properties of Boron Nitride. Hexagonal BN is a promising candidate for use as a microwave window material. The window will reach temperatures in the neighborhood of 3000 K for short periods of time. It is therefore important to know the physical and chemical properties of BN in this range. The focus of this project is on the electrical d.c. conductivity of BN and on its dielectric behavior when subjected to microwave radiation at very high temperatures.

The first four months (Jan.-April) have been used to explore various experimental approaches to the measurement of the electrical resistance and the temperature in the presence of large electrical and thermal leaks. At the same time, computer programs were developed to calculate the dielectric parameters of BN in the low-GHz frequency range.

Electrical Conductivity

Sample Configuration

Although hexagonal boron nitride possibly is the best electrical insulator known up to 2000 °C, above this temperature the conductivity rapidly increases to values in excess of $10^{-3} \text{ ohm}^{-1} \text{ cm}^{-1}$. This conduction probably is intrinsic in nature. In order to prevent degradation and nonstoichiometry due to loss of nitrogen, which could cause large extrinsic conduction, it is necessary to perform high temperature

measurements in nitrogen gas. According to the literature [1], the equilibrium vapor pressure of N_2 over BN is 0.1 atm at 2265 °C, 1.0 atm at 2505 °C, and 3.0 atm at 2626 °C.

As stated in the first Progress Report (May 1982), above 2000 °C one observes a leakage conductance--probably due to gas conduction--which reaches values comparable to that of the BN sample. Consequently, it is necessary to devise an experimental arrangement which avoids the problem of the large leakage conduction path. One solution is a geometry in which the sample is isolated from the nitrogen gas (Scheme #1). Another arrangement is a guard ring configuration whereby the sample and sample leads are electrically shielded from the large leak-currents to ground (Scheme #2).

Scheme #1

For this approach, the insulation of the sample from the gas is provided largely by boron nitride at a lower temperature, as shown in Fig. 1. The actual BN sample is confined mainly to the volume between the tungsten electrodes. This "sample" is part of a BN cylinder which is positioned at the access opening of the lowest of the upper eight heat shields. Above this level, the temperature drops rapidly. Consequently, the boron nitride cylinder above the top electrode has a high resistance and contributes little to the conduction path. The bottom tungsten electrode is held in place by a threaded BN plug. Due to this geometry, it is difficult to translate the resistance, R , of the sample into resistivity. However, a rough estimate indicates that the numerical value of the resistivity is of the order of 1.5 R and certainly does not exceed 2.0 R .

In the first run, the temperature of the sample was measured by a W(3% Re) vs. W(25% Re) thermocouple attached to the top electrode. The temperature in the center of the furnace was determined by means of a W(5% Re) vs. W(26% Re) thermocouple introduced through the bottom of the furnace and by means of a pyrometer sighted through a viewing port on the side of the furnace. The readings of the pyrometer and of the thermocouple in the center of the furnace agreed within 40-50 °C up to 2200 °C; beyond this temperature, the latter was retracted from the furnace.

The sample used in the first run was cut from a small block of high purity boron nitride (HPBN-TS1251; Union Carbide). The temperature at the center of the furnace was raised to a maximum of 2480 °C, as indicated by the pyrometer. At this point, the sample temperature lagged behind considerably: $T = 1850$ °C was the highest reading. All measurements were made with an N_2 pressure of 1.0 atm. Results of this first run are shown in Fig. 2.

In order to raise the sample temperature in the next run, the boron nitride cylinder was made somewhat longer, such that the top electrode was located 1.0 cm below the lowest horizontal heat shield. This made it possible to reach a higher sample temperature of 2021 °C when the maximum temperature in the center of the furnace was 2410 °C, as indicated by the pyrometer. Conductivity measurements were made at three different pressures of the nitrogen gas: 1.0, 2.0, and 3.0 atm. Results of this second run are also presented in Fig. 2.

Electrical measurements during the first run were terminated when the tungsten wire failed between the sample and the BN insulator which

extended through the viewing port into the hot zone of the furnace (see Fig. 1). An attempt to push the furnace temperature above 2500 °C resulted in considerable effervescence and partial destruction of the end of the BN insulator. The second run was terminated at a furnace temperature of about 2500 °C when the wires of the lower thermocouple on the sample broke.

Two materials failure modes are indicated by these results. The first mode is the decomposition of BN at temperatures in excess of 2500 °C under a pressure of 1.0 atm of N₂. This observation is in agreement with the literature value of 2505 °C [1]. No decomposition of BN was evident, as expected, in the second run when the pressure was increased to 3.0 atm of N₂.

The second mode appears to occur through a reaction between the tungsten wires and the BN parts. After each experiment, the BN sample and insulators were inspected radiographically. Radiographs of sample and insulators are shown in Fig. 3. The multiple breaks and balling up of the tungsten wire suggest that the wire failed by forming a liquid phase. This is in agreement with the W-B phase diagram [2]: W₂B₅ melts at 2365 °C and excess boron lowers the melting temperature to ~ 2000 °C (Fig. 4). When the amount of tungsten exceeds the BN present (W > 50 percent), the melting points of the W-B compounds and solid solutions are 2600 °C or higher. Consequently, more massive pieces of tungsten (rods and discs), in contact with BN, should not be subject to disintegration if one keeps the temperature below ~ 2600 °C.

Scheme #2

In this arrangement (Fig. 5), the sample is located near the center of the furnace suspended from a tungsten tube, which acts as an elec-

trical shield. A "lip" at the bottom of this shield functions as the guard ring. The sample is provided with two tungsten discs; these are the electrodes for measuring the resistance of the BN sample. All these parts are held together by two BN rings, threaded on the cylindrical sample.

The sample resistance, R_x , is measured by means of an OP-AMP circuit [3]; R_x is deduced from the following expression:

$$\frac{R_x}{E_i} = - \frac{R_f}{E_o} ,$$

where: R_x = sample resistance,

E_i = applied voltage,

R_f = feedback resistance, and

E_o = output voltage.

The OP-AMP circuit plays the role of a very sensitive nulling device (OP-AMP A). The circuit is strictly d.c.; OP-AMP's B and C are filters to prevent any intrusion from low frequency a.c. sources.

The shield and the main furnace are at ground potential; point P, between R_x and R_f , is only a few microvolts above ground. Consequently, no leakage can occur between the top lead to the sample and its surroundings. Possible leakage paths from the lower sample lead to the grounded furnace body are not part of the main circuit (as long as E_i does not vary) and, hence, are not being measured.

Conductivity measurements on several BN and BN-containing samples are in progress.

Theory of Microwave Window Transmission

1. General Considerations

Under the conditions of atmospheric reentry, the ARV microwave window material is subjected to most severe environmental conditions. The very high outer surface temperature leads to ablation of the material and the flow of a heat pulse into the window. The temperatures reached are sufficiently high that the thermal generation of conducting carriers is sufficient to cause significant microwave absorption (and reflectance) in many candidate materials. The processes induced by reentry are so rapid that the temperature distribution and carrier concentrations are nonuniform.

Complete modeling of the transmissivity of a microwave window would require a knowledge of the ablation rate, the temperature profile, and the enhancement of electrical conductivity at all times during the reentry and also the effects of the external plasma of ionized gases. In this part of the project, we address the more limited question of finding the effects of the intrinsic conduction processes on the applicability of boron nitride for window use. We devote the principal effort to the calculation of the microwave transmission through window material at high temperatures, with a steep temperature gradient and with thermal excitation of conductivity.

The calculations we perform are based on the model illustrated in Fig. 6. Microwave radiation is incident from the left. Significant reflection (to be calculated) occurs; the radiation in the window undergoes multiple internal reflections and a transmitted wave emerges, propagating to the right. As the window ablates, the thickness of the

window decreases, leading to interference oscillations in the transmissivity. An additional complication is that, for the conductivities and the microwave frequencies that are used, the electromagnetic skin depth and the wavelength in the window are smaller than the thickness of the hot layer at the outer surface. Thus, we must solve the problem of electromagnetic radiation passing through a medium with parameters that are changing slowly with respect to the wavelength in the medium.

Because of the immediate availability of suitable computer programs, we felt that direct numerical calculation was the simplest way to treat this problem correctly. Approximations utilizing the WKBJ [4] method could have been used, but computer usage would probably have been necessary anyway.

2. Method of Calculation

The propagation of an electromagnetic wave in a medium having a spatially varying electrical conductivity is described by Maxwell's equations. If we assume that the material variation is a function of one coordinate, z ; i.e., that the medium is stratified and that the radiation propagate is parallel to the z -direction, then the wave is described by the pair of coupled linear differential equations:

$$\frac{dE}{dz} = -ikZ_0 H \quad , \quad (1.a)$$

$$Z_0 \frac{dH}{dz} = -ik\epsilon(z)E \quad . \quad (1.b)$$

In the above, E and H are the electric and magnetic field intensities, which are mutually perpendicular, and both are transverse to the z -

direction. Sinusoidal monochromatic fields of angular frequency, ω , are assumed; the convention for the time variation is by use of the understood factor, $e^{-i\omega t}$. Z_0 is the impedance of free space, and k is the wave vector equal to the reciprocal wavelength times 2π . The complex dielectric constant, ϵ , is given by,

$$\epsilon(z) = \epsilon_0 \kappa_e + i\sigma(z)/\omega \quad , \quad (2)$$

where ϵ_0 is the permittivity of free space, κ_e is the relative dielectric permittivity of the window material, and $\sigma(z)$ is the spatially varying electrical conductivity.

In the above, SI units are used exclusively. The numerical solutions of Eqs. (1.a) and (1.b), subject to specially imposed boundary conditions, enable one to compute the characteristic matrix of the material. By using standard methods [5], which we shall not reproduce here, the transmissivity and reflectivity of material of arbitrary dielectric profile can be obtained once the characteristic matrix is computed.

The problem is thus completely specified if we give the temperature profile and the dependence of conductivity on temperature. For the temperature profile, we use the values reported by the TRW Corporation [6] on data for hot pressed boron nitride. The profile is given approximately by the exponential form,

$$T = T_0 e^{-\alpha z} \quad , \quad (3)$$

where $\alpha = 0.216 \text{ cm}^{-1}$ is the decay constant and T_0 is the surface temperature. For the conductivity of BN, we use the loss tangent data compiled by Hathaway et al. [7]. The loss tangent, $\tan \sigma$, is related to the conductivity by,

$$\tan \sigma = \frac{\sigma}{\epsilon_0 \kappa_e \omega} . \quad (4)$$

Here, κ_e is the dielectric constant of BN, taken as 3.1. Their data [7] are fitted well by the Arrhenius form,

$$\sigma(T) = \sigma_0 e^{-\Delta/kT} , \quad (5)$$

where the prefactor, $\sigma_0 = 1.63 \times 10^3 \Omega^{-1} \text{ cm}^{-1}$, and $\Delta = 3.08 \text{ eV}$, are consistent with a band gap of 6.0 eV. For a surface temperature of 3150 K, the conductivity at the surface would be $2.0 \Omega^{-1} \text{ cm}^{-1}$; very close to the values extrapolated from the measurements of this report (see Fig. 2).

3. Results of the Calculations and Discussion

In Fig. 7 through Fig. 10, we show the calculated transmissivities of boron nitride windows as a function of thickness for a range of surface temperatures. The curves have common characteristics which we now shall describe. The transmissivity falls very rapidly as window thickness increases from zero. The curves all show that the principal effects of the conduction processes occur in the outer one or two millimeters of the hot face. All curves show a near-sinusoidal oscillation with thickness, which is characteristic of the multiple internal reflec-

tions that occur in the cool part of the window. The percent transmissivity increases as the surface temperature is reduced, to as high as nine percent for $T_0 = 2850$ K.

In the various cases studied, the reflectivity varied from 80 percent to 85 percent, and the absorptivity varied from 10 percent to 15 percent. This demonstrates the mirror-like properties of the outer hot surface of the window.

The oscillatory dependence upon thickness suggests that one could observe, at 10 GHz, a series of transmissivity peaks as the ablation process proceeds. We consider the peak values as the maximum obtainable transmissions, for if the window were coupled to a continuously tuned cavity the interference cancellation could be removed.

The conclusion to be drawn from these calculations is that, at the considered reentry conditions, the transmissivity of intrinsic boron nitride windows at 10 GHz will have an upper limit of 10 percent; i.e., the one-way loss will be 10 dB or greater. Some improvement could be obtained by altering the material composition and form in such a way as to reduce the surface temperature or to increase the temperature gradient. Calculations of this type should be extended to materials, such as woven BN-SiO₂ fiber composites and other candidate materials. Also, an improved theory of the temperature profile should be developed.

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Figure Captions

1. Boron Nitride sample with internal electrodes (Scheme 1):
 - A,B--tungsten electrodes
 - T--W(3Re) vs. W(25Re) thermocouple
 - H--heater
 - S--radiation shields
2. Resistance of BN as a function of reciprocal temperature:
 - o--Run I
 - x--Run II; $P_{N_2} = 1.0$ atm
 - o--Run II; $P_{N_2} = 3.0$ atm
3. Radiographs of BN sample.
4. Phase Diagram of W-B system.
5. Schematic of sample holder, furnace, and OP-AMP circuit (Scheme 2).
6. Illustration of the model for calculating the microwave transmissivity of BN windows.
7. Calculated transmissivity of BN microwave windows as a function of thickness for 1.0 GHz radiation and a surface temperature of 3150 K (2877 °C).
8. Calculated transmissivity of BN microwave windows as a function of thickness for $f = 10.0$ GHz and $T = 3150$ K (2877 °C).
9. Calculated transmissivity of BN microwave windows as a function of thickness for $f = 10.0$ GHz and $T = 3000$ K (2727 °C).
10. Calculated transmissivity of BN microwave windows as a function of thickness for $f = 10.0$ GHz and $T = 2850$ K (2577 °C).

FIGURE 1

BORON NITRIDE SAMPLE WITH
INTERNAL ELECTRODES

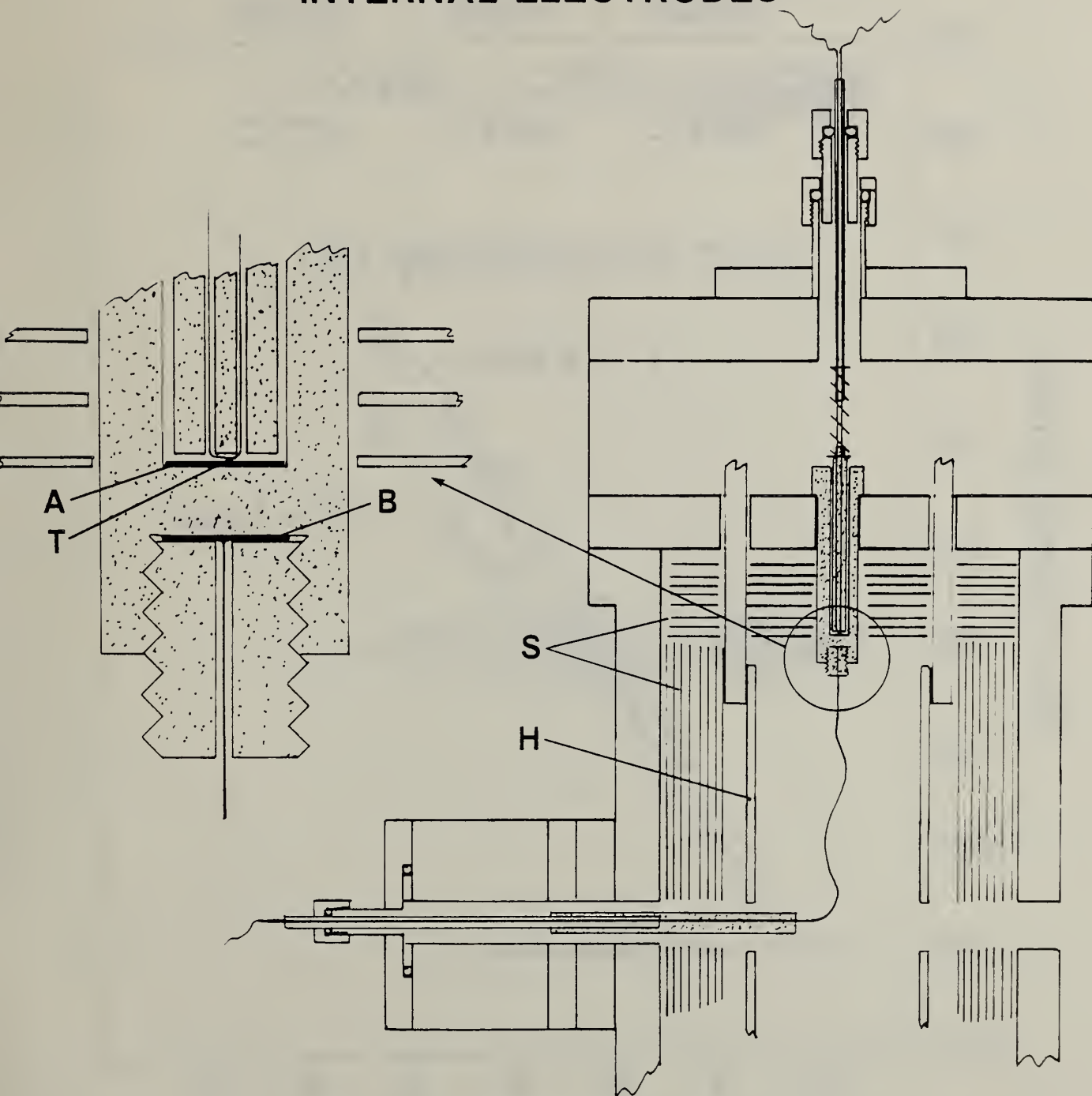


FIGURE 2

RESISTANCE OF BN vs RECIPROCAL TEMPERATURE

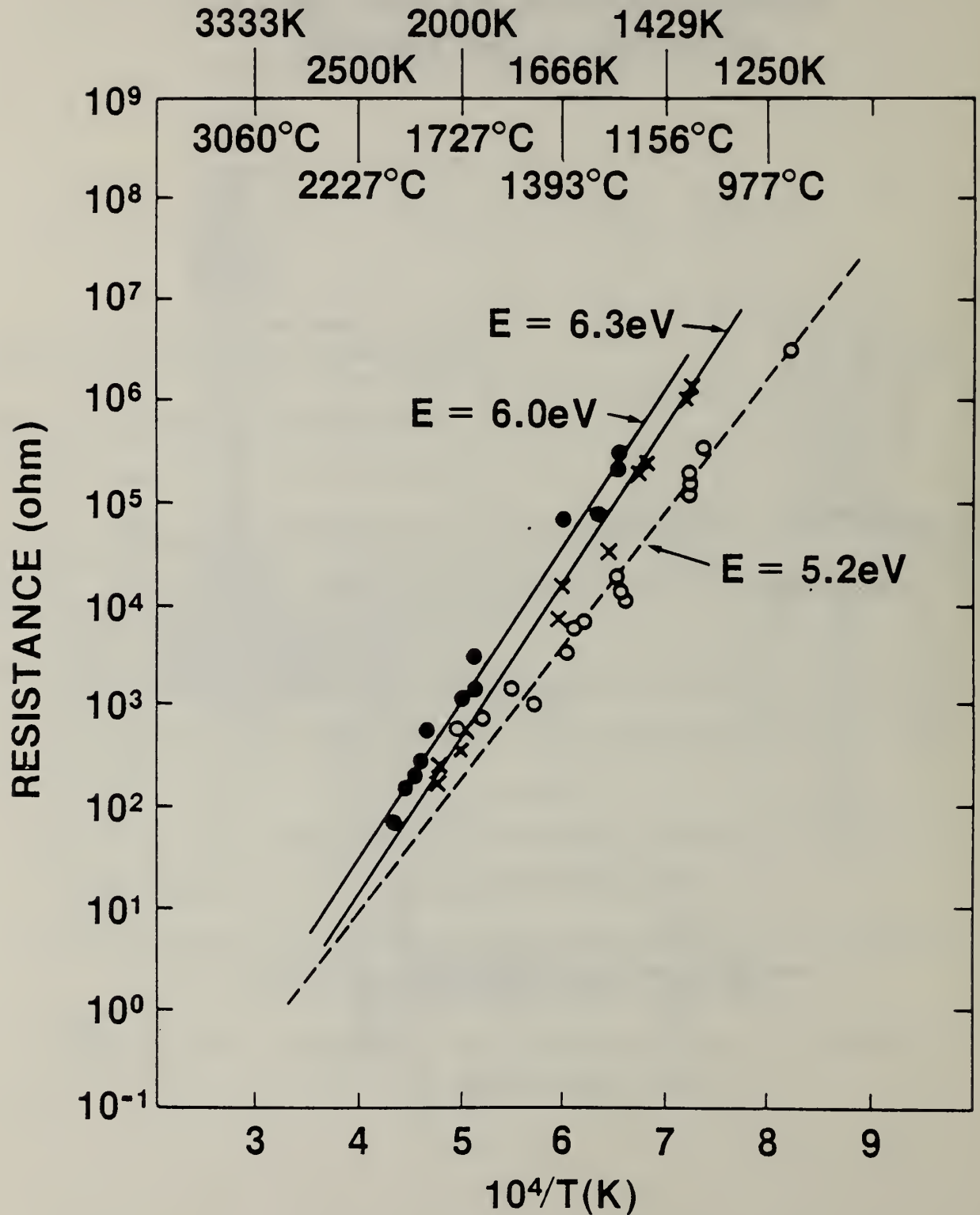
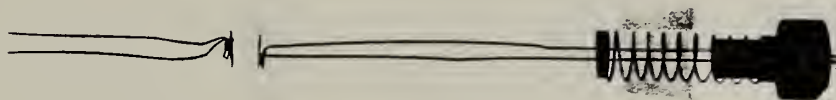
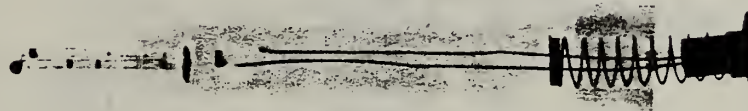


FIGURE 3
RADIOGRAPHS OF BN SAMPLE

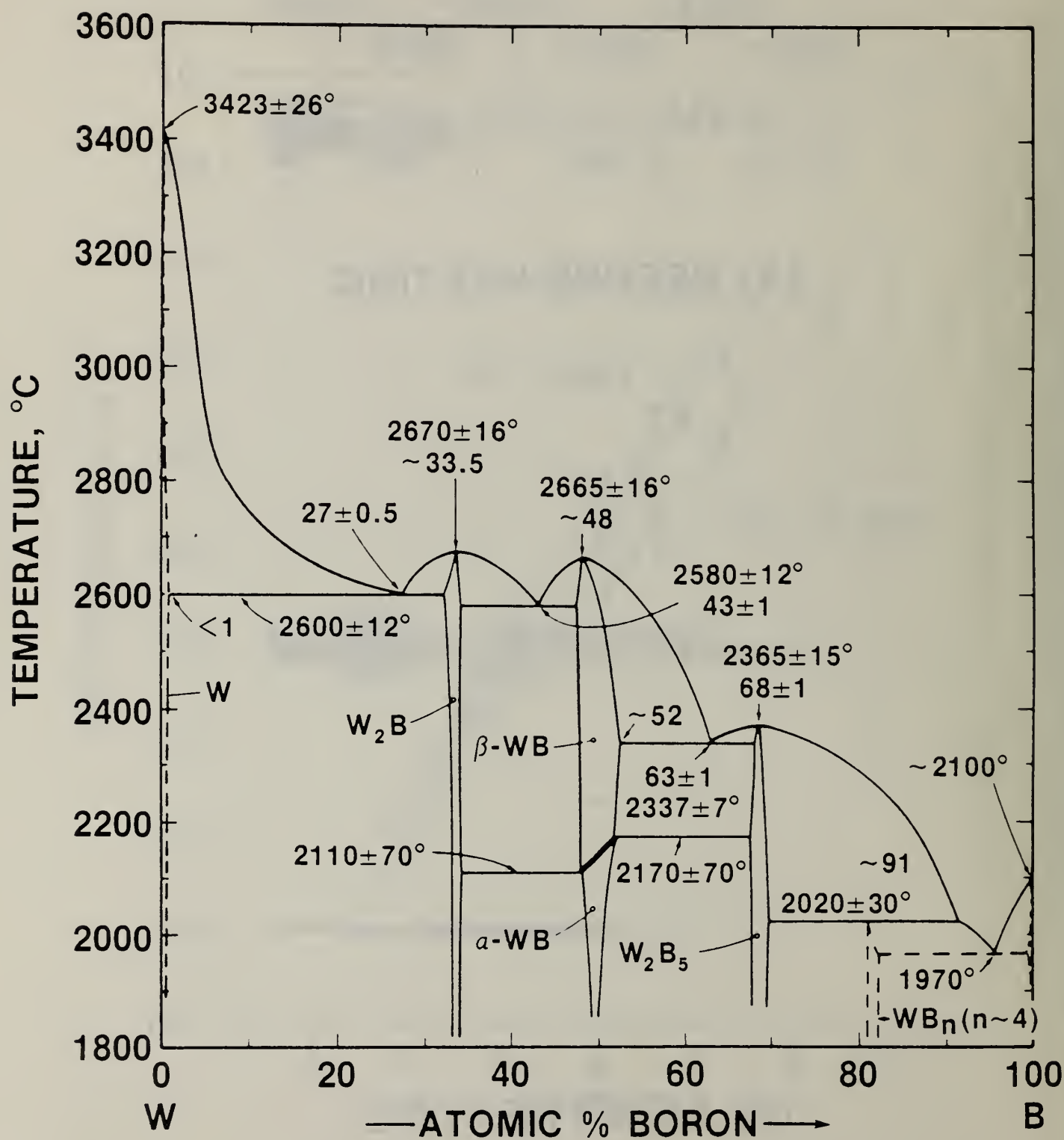


(A) BEFORE HEATING



(B) AFTER HEATING

FIGURE 4
PHASE DIAGRAM OF W-B SYSTEM



SCHEMATIC OF SAMPLE HOLDER, FURNACE, AND OP-AMP CIRCUIT

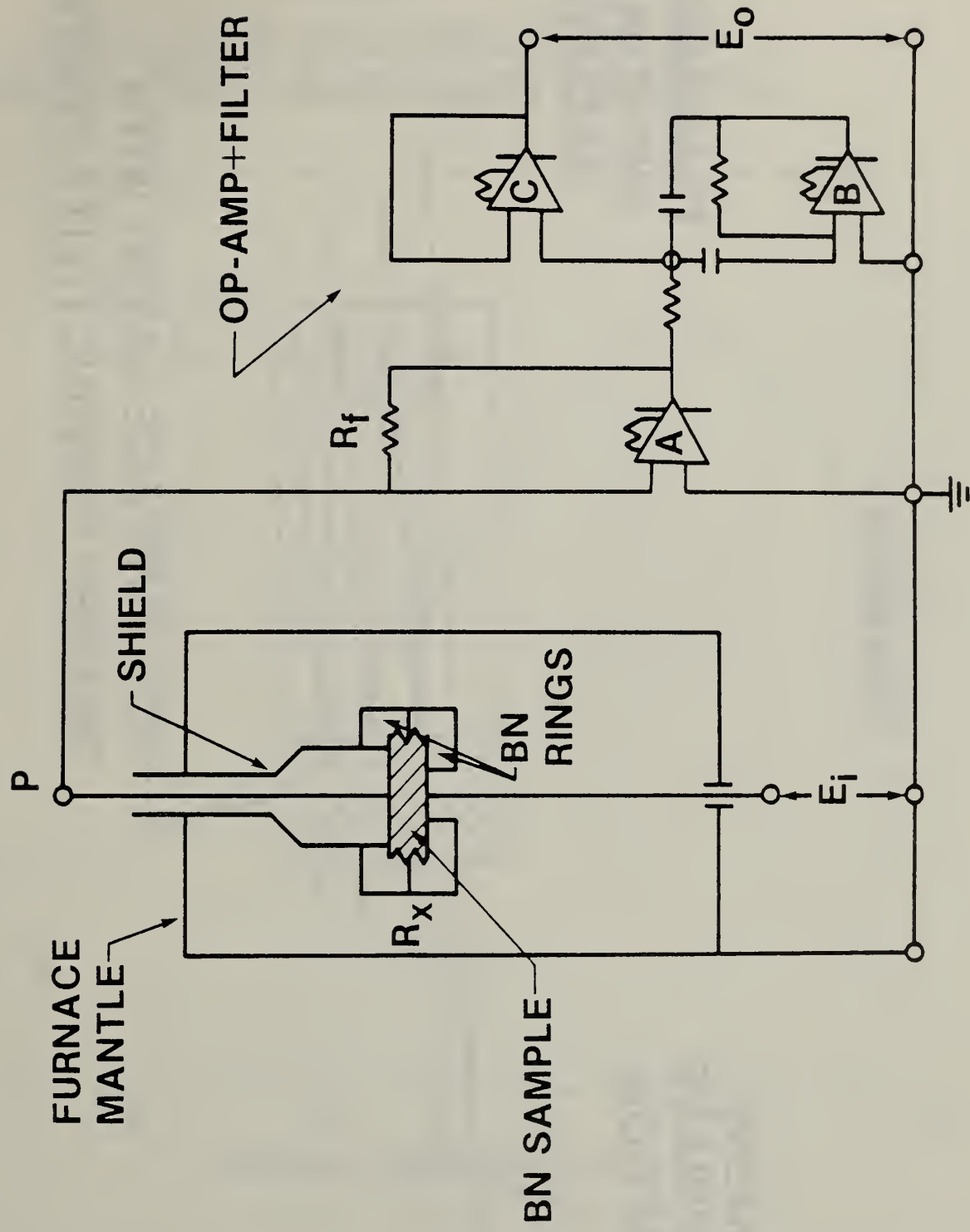


FIGURE 6

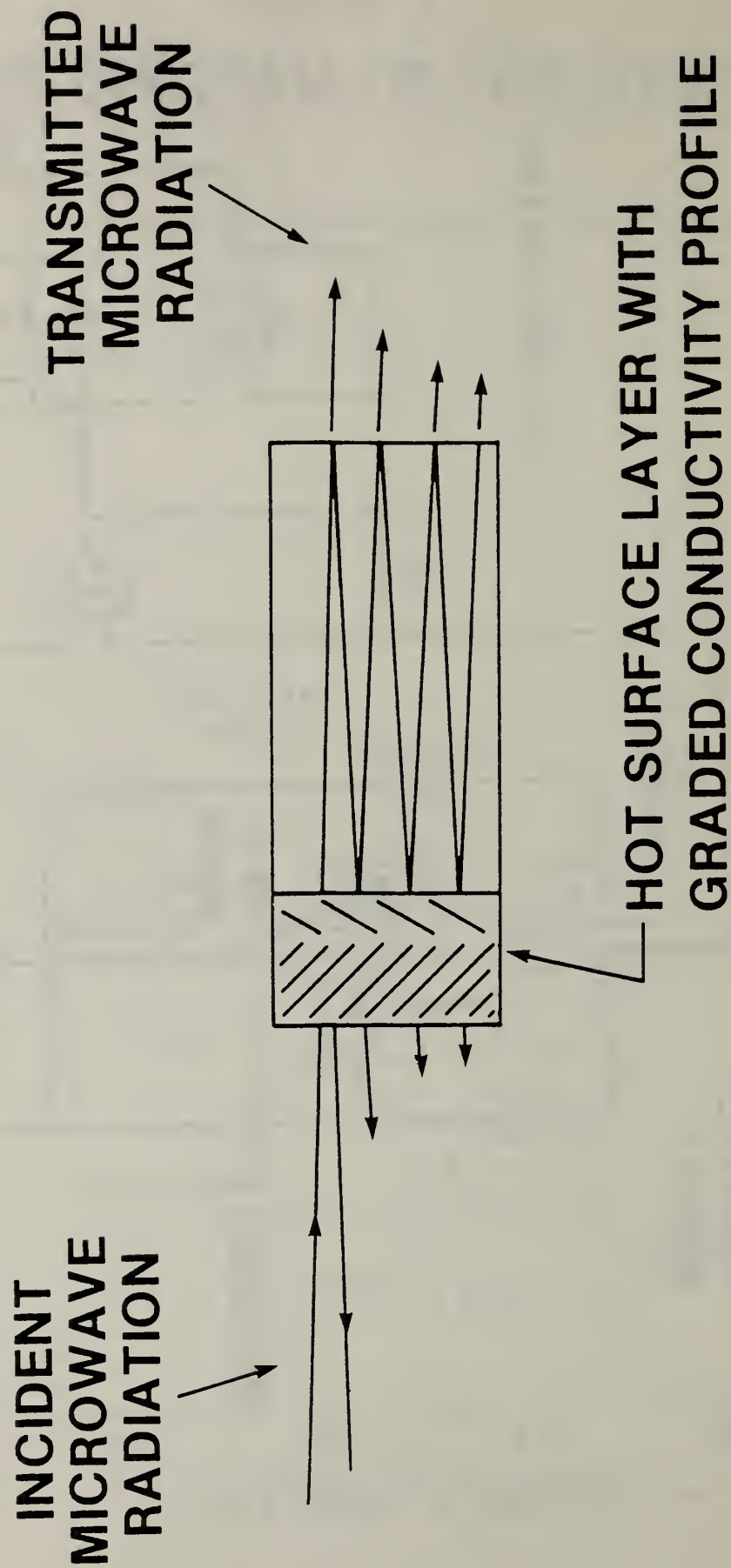


FIGURE 7

CALCULATED TRANSMISSIVITY OF
BORON NITRIDE MICROWAVE WINDOWS

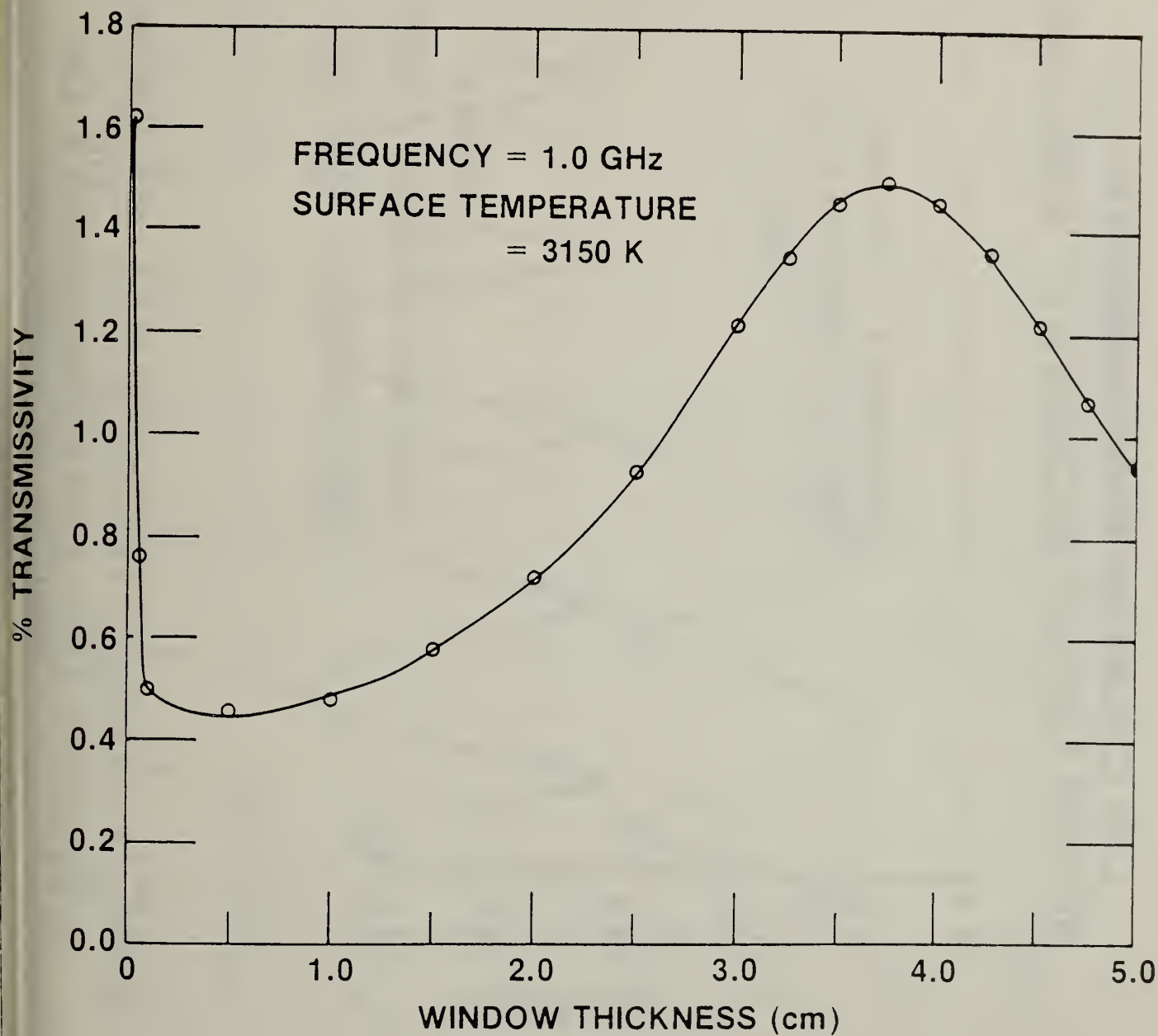
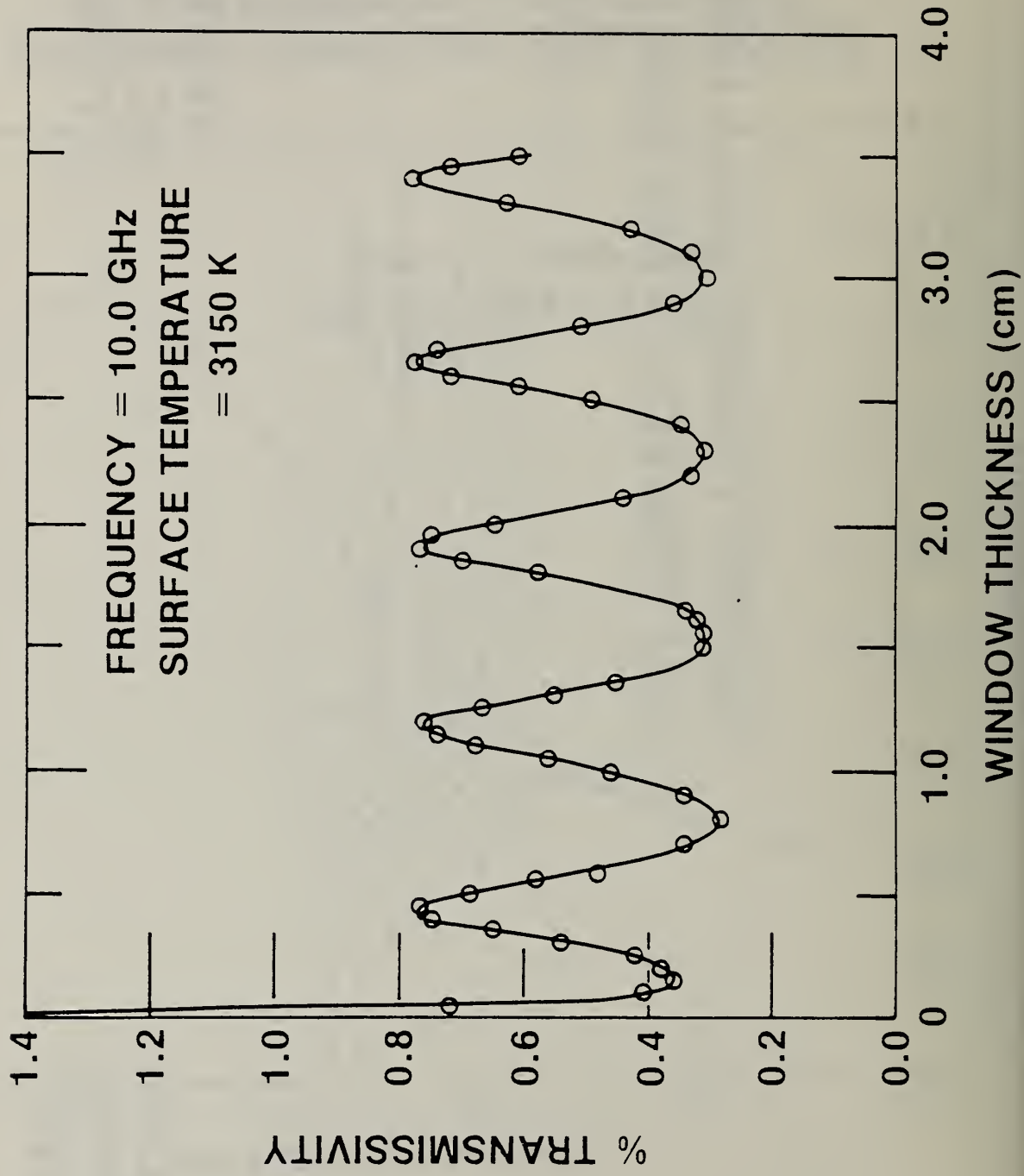


FIGURE 8

CALCULATED TRANSMISSIVITY OF
BORON NITRIDE MICROWAVE WINDOWS



CALCULATED TRANSMISSIVITY OF BORON NITRIDE MICROWAVE WINDOWS

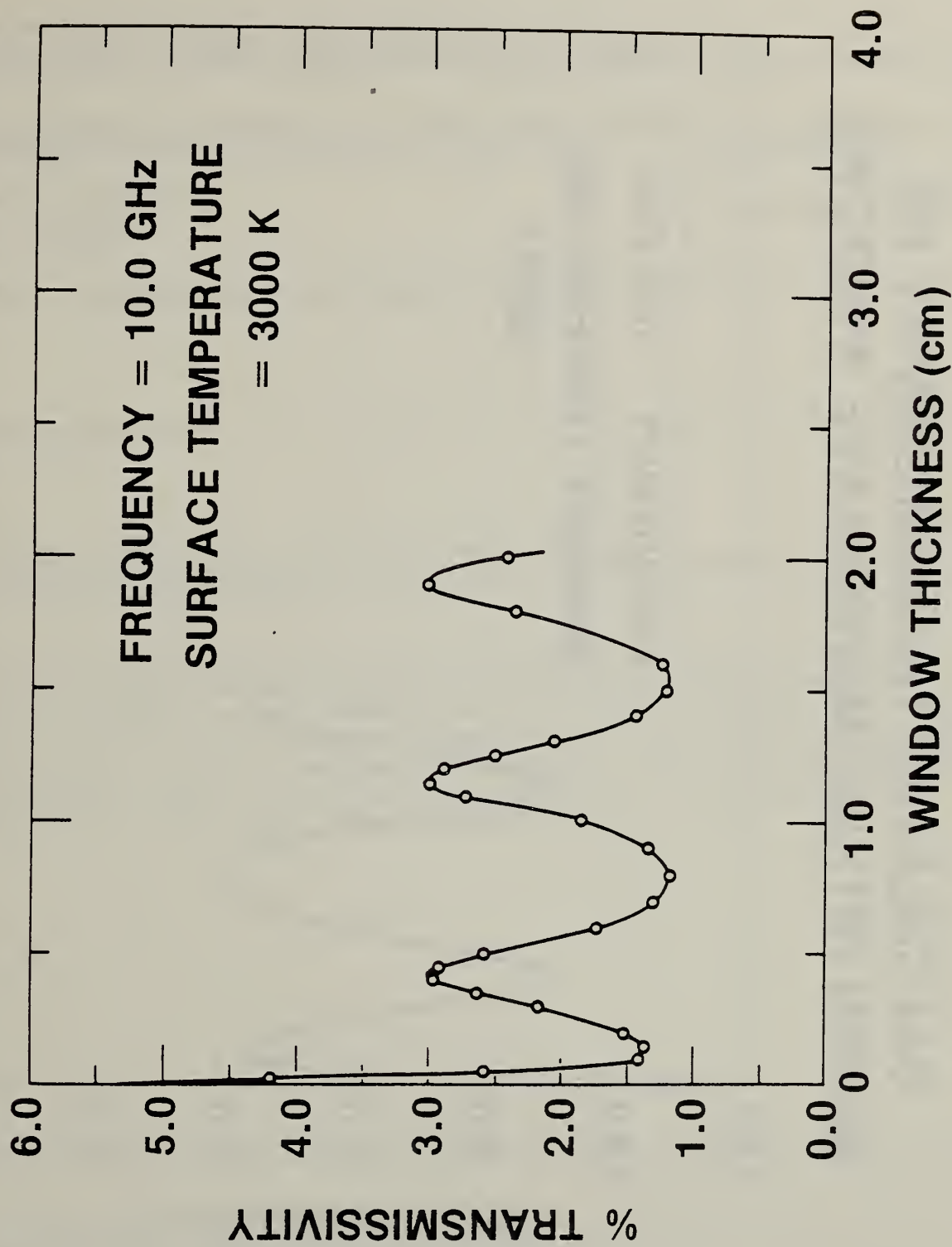
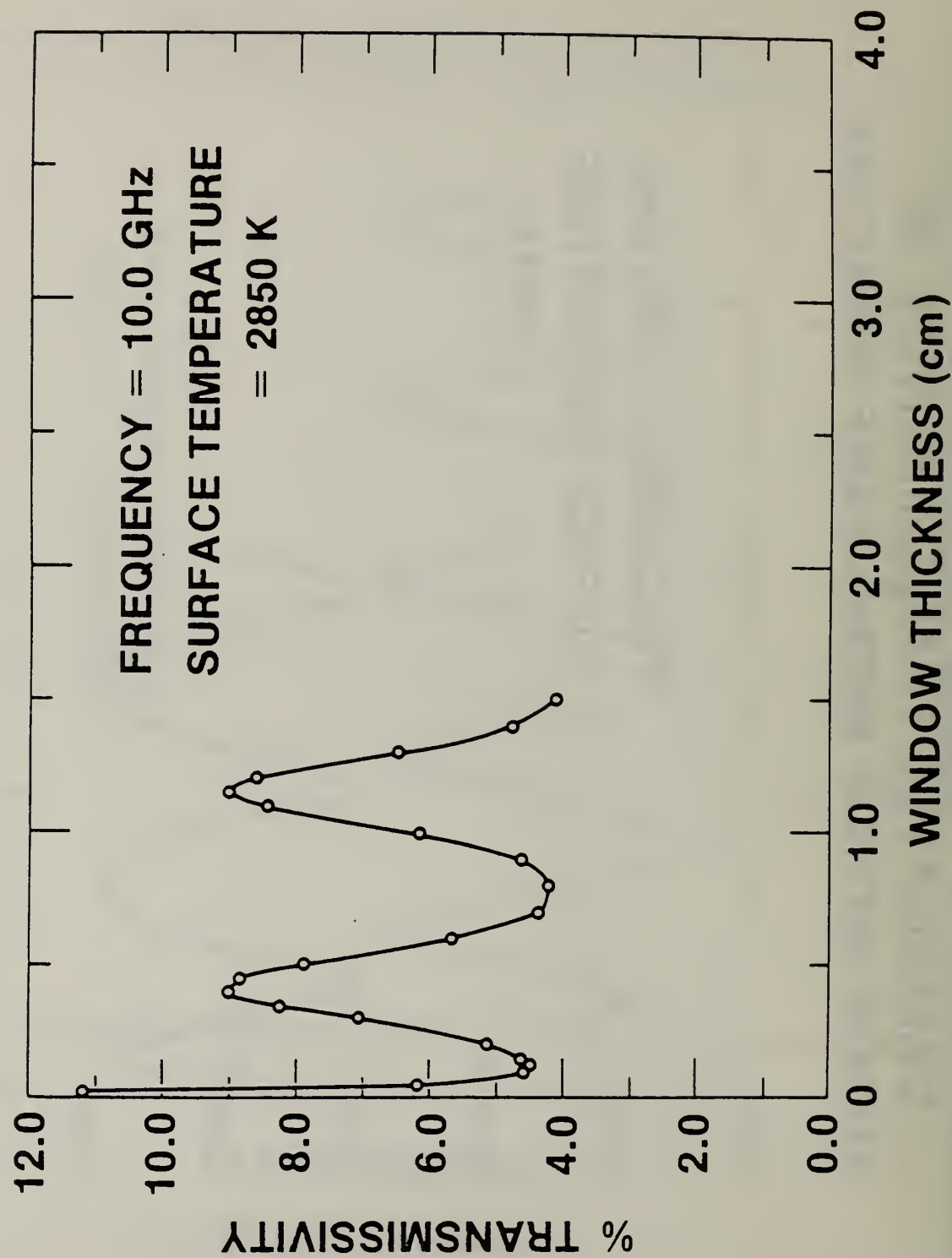


FIGURE 10
CALCULATED TRANSMISSIVITY OF
BORON NITRIDE MICROWAVE WINDOWS



U.S. DEPT. OF COMM. BIBLIOGRAPHIC DATA SHEET (See instructions)		1. PUBLICATION OR REPORT NO. NBSIR 83-2646	2. Performing Organ. Report No.	3. Publication Date January 1983
4. TITLE AND SUBTITLE "Characterization of ARV Antenna Window Material" (Progress Report No. 2, October 1982; Defense Nuclear Agency)				
5. AUTHOR(S) H. P. R. Frederikse, A. L. Dragoo, A. H. Kahn, and W. R. Hosler				
6. PERFORMING ORGANIZATION (If joint or other than NBS, see instructions) NATIONAL BUREAU OF STANDARDS DEPARTMENT OF COMMERCE WASHINGTON, D.C. 20234			7. Contract/Grant No.	8. Type of Report & Period Covered
9. SPONSORING ORGANIZATION NAME AND COMPLETE ADDRESS (Street, City, State, ZIP)				
10. SUPPLEMENTARY NOTES <input type="checkbox"/> Document describes a computer program; SF-185, FIPS Software Summary, is attached.				
11. ABSTRACT (A 200-word or less factual summary of most significant information. If document includes a significant bibliography or literature survey, mention it here) Boron nitride is a major candidate for use as a microwave window material. To assess its feasibility for this purpose, it is essential to know the electrical and dielectric properties at high temperatures. This report discusses the experimental approach to electrical conductivity measurements above 1800 °C and presents some initial results. At the same time, computer calculations have been performed which have yielded values of the reflectivity, absorption, and transmission of boron nitride in the temperature range 2500-3000 °C.				
12. KEY WORDS (Six to twelve entries; alphabetical order; capitalize only proper names; and separate key words by semicolons) Boron nitride; Calculated transmissivity; Electrical conductivity; High temperatures; Microwave radiation				
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